Transport in coupled graphene-nanotube quantum devices

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We report quantum transport measurements on carbon nanotube quantum dot devices with graphene charge detectors and a nanotube contacted by graphene source and drain leads. Individual charging events in the nanotube quantum dot lead to conduction changes of up to 20% in the graphene nanoribbon acting as a charge detector. In a graphene-contacted nanotube device we observe Coulomb blockade leading to the conclusion that tunneling barriers with rates in the low GHz regime are present at the graphene carbon nanotube interfaces.

Carbon nanomaterials, such as graphene and carbon nanotubes (CNTs) attract increasing interest mainly due to their promises for flexible electronics, high-frequency devices and spin-based quantum circuits [1–3]. Both materials consist of sp²-bound carbon and exhibit unique electronic properties resulting in the suppression of direct backscattering, high carrier mobilities and low intrinsic spin noise. In particular the weak hyperfine interaction makes graphene and CNTs interesting host materials for quantum dots which promise the implementation of long-living spin qubits [4]. Up to the present state quantum dots (QDs) and double quantum dots have been demonstrated successfully in both carbon nanomaterials. In contrast to graphene, the fabrication of ultra-clean few-carrier QDs with well-defined spin states in quasi one-dimensional (1-D) carbon nanotubes is well established [5–7]. A comparable quality in graphene is not yet reached, which is mainly due to its gap less band structure making it hard to controllably confine electrons and holes. State-of-the-art graphene QDs are therefore either based on nanoribbons [8, 9] or etched islands [10–14] and in both cases edge roughness and disorder is dominating their properties. However, in contrast to quasi 1-D nanotubes, the 2-D nature of graphene makes it easy to integrate lateral graphene gates and in-plane charge sensors [15] which are both important for the control and readout of QD states. Here, we present quantum devices based on both graphene and carbon nanotubes, which combine the advantages of the two carbon allotropes. In particular, we discuss (i) a carbon nanotube QD with a capacitively coupled graphene nanoribbon acting as electrostatic gate and charge detector and (ii) a nanotube QD with both source and drain graphene leads. Thus, a quantum dot device exclusively built of two different carbon allotropes. Interestingly, both materials are also technologically highly relevant in the frame of the International Technology Roadmap for Semiconductors [16] and first proposals for coupled graphene-nanotube devices have been reported just recently [17].

The fabrication process is based on chemical vapor deposition (CVD) growth of carbon nanotubes and subsequent deposition of mechanically exfoliated natural graphite. In Figs. 1(a) to 1(c) we show the three

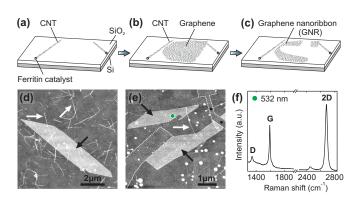


FIG. 1. (color online) (a),(b) and (c) Schematic illustrations of the three main fabrication steps. (d) Scanning force microscopy (SFM) image of a SiO₂ substrate with CVD grown CNTs (white arrows) and subsequently deposited graphene (black arrows). (e) SFM image of a similar sample after the dry etching process. White dashed lines indicate areas exposed to an ${\rm Ar/O_2}$ plasma which results in etching both carbon materials as indicated by the black points. (f) Raman spectrum of the graphene flake at the position indicated by the green dot in panel (e). The spectrum shows clear characteristics of single-layer graphene.

main fabrication steps for making all-carbon graphenenanotube devices. As a first step single-walled carbon nanotubes are grown on 290 nm SiO₂ on highly p-doped Si substrates by CVD using a Ferritin-based iron catalyst method [18]. The single-walled carbon nanotubes have a diameter of around 1.5-2 nm and are up to several micrometers in length. In a next step, graphene is deposited on these pretreated substrates by mechanical exfoliation of natural graphite [19]. Figs. 1(d) and 1(e) show scanning force microscopy (SFM) images of two examples of the deposition process. Due to the stochastic nature of the method a great variety of different configurations is obtained. For example, the graphene flake shown in Fig. 1(d) (black arrow) lies on top or nearby of a number of individual CNTs (white arrows) whereas in Fig. 1(e) we show an example where a nanotube (white arrow) is contacted by two graphene flakes (black arrows). A crucial parameter for the success of this stochastic fabri-

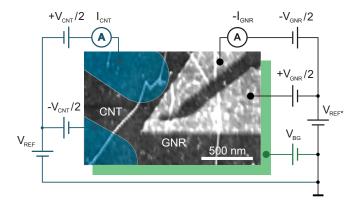


FIG. 2. (color online) Scanning force micrograph of a carbon nanotube (CNT) quantum dot device with a nearby structured graphene nanoribbon (GNR) for charge sensing. The schematic illustration shows the used measurement circuit and the applied voltages. For detailed information see text.

cation process is the density of grown CNTs which can be well controlled by the Ferritin-based CVD process [18]. In this study we used a density of approx. 1-2 CNTs per μ m². In Fig. 1(f) we show a Raman spectrum taken at the position indicated by the green dot in Fig. 1(e). From the full width at half maximum of the 2D peak of 34 cm⁻¹ and the relative intensity of the 2D and G peak of $I(G/2D)\approx 0.65$, we conclude that the graphene is of single-layer nature [20, 21] (laser excitation of 532 nm). A Raman spectrum of the bottom graphene flake at the position of the intersecting CNT reveals a bilayer flake (not shown). The contacted CNT structure consists of two segments. Most likely the upper part (segment) is a small bundle of single-walled CNTs whereas the lower part (white arrow in Fig. 1(e)) is an individual singlewalled CNT. This conclusion is supported by a comparison of the SFM profiles of the single-layer graphene and the lower carbon nanotube segment, as well as the high accuracy of the Ferritin-based CVD growth process [18]. The third fabrication step consists of electron beam lithography (EBL) followed by an Ar/O₂ based dry etching step for (i) structuring graphene and (ii) removing unwanted CNTs. In Fig. 1(e) we highlight areas (dashed lines) where graphene and CNTs have been successfully removed (see black points). Finally, we used EBL, metal evaporation (5 nm Cr/50 nm Au) and lift-off for contacting the devices.

Fig. 2 shows a SFM image of an all-carbon device consisting of a carbon nanotube lying in the close vicinity to an etched graphene nanoribbon (GNR) which acts as a charge detector (CD). By Raman spectroscopy we identify the nanoribbon to be of bilayer nature (not shown) and following the previous argument we conclude that the nanotube is a single-walled carbon nanotube. Both carbon nanostructures are separated by roughly 150 nm, the nanoribbon has a width of around 100 nm and the CNT quantum dot is defined by two metal electrodes (indicated in blue) which are separated by 350 nm. As

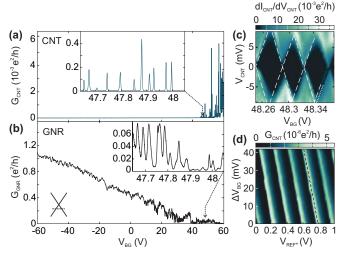


FIG. 3. (color online) (a),(b) Back gate characteristics of the carbon nanotube QD (a) and the graphene nanoribbon (b). Both measurements were recorded at a source-drain bias voltage of $V_{\rm CNT}=V_{\rm GNR}=0.5~\rm mV$ and $V_{\rm REF}=0~\rm V$. The upper inset in panel (a) shows Coulomb blockade resonances of the CNT QD as a function of $V_{\rm BG}$. The inset in panel (b) highlights the conductance resonances in the GNR in the same range of $V_{\rm BG}$. (c) Differential conductance on the CNT QD. The white dotted line indicates an excited state with an energy of $\Delta=0.9~\rm meV$. (d) Conductance of the CNT as function of $\Delta V_{\rm BG}=V_{\rm BG}-13.63~\rm V$ and $V_{\rm REF}*$ at $V_{\rm CNT}=20~\rm mV$. The black dashed line highlights the relative lever arm $\alpha_{\rm BG,GNR}$.

illustrated in Fig. 2 we apply a symmetric bias voltage ($V_{\rm CNT}$ and $V_{\rm GNR}$ respectively) to both structures. The overall Fermi level can be tuned by the back gate voltage $V_{\rm BG}$ applied to the highly doped Si substrate. Additionally, we can use the CNT (GNR) as a lateral gate for the GNR (CNT) by applying a reference potential $V_{\rm REF}$ ($V_{\rm REF}$ *). All presented measurements were performed in a pumped $^4{\rm He}$ -cryostat at a base temperature of T \approx 1.5 K using low-frequency lock-in techniques.

Figs. 3(a) and 3(b) show the back gate characteristics i.e. the conductance as function of $V_{\rm BG}$ of both, the CNT (Fig. 3(a)) and the graphene nanoribbon (Fig. 3(b)) for a constant bias voltage $V_{CNT}=V_{GNR}=0.5$ mV. The CNT reveals a semiconducting behavior with a large band gap resulting in an extended BG region of suppressed current (-60 V< V $_{\rm BG}$ <40 V). A high resolution measurement performed at the edge of this gap (see inset in Fig. 3(a)) exhibits reproducible, well resolved and sharp Coulomb peaks. The peak width of the sharpest resonances is given by the electron temperature [22] which can be extracted to be 2 K. In Fig. 3(c) we show so-called Coulomb diamond measurements i.e. the differential conductance dI_{CNT}/dV_{CNT} plotted as function of V_{BG} and V_{CNT} . From the extent of suppressed current of the diamonds in bias (V_{CNT}) direction we can extract an addition energy of 6-9 meV of the CNT quantum dot and a BG lever arm, $\alpha_{\rm BG}=0.22$. The change in differential conductance parallel to the diamond edges is attributed

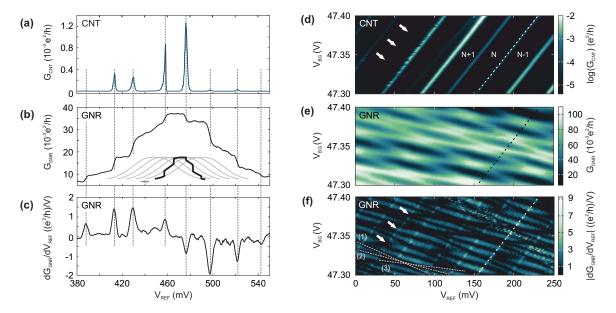


FIG. 4. (color online) (a) QD conductance and (b) GNR conductance as a function of V_{REF} . A bias voltage of $V_{CNT}=V_{GNR}=0.5$ mV was applied to both structures. The inset in (b) shows a schematic of the charge detecting mechanism. (c) Derivative of (b) showing well pronounced peaks at every charging event. (d) Dependence of the CNT QD conductance on V_{BG} and V_{REF} measured at $V_{CNT}=0.5$ mV on a logarithmic scale. Periodic Coulomb blockade resonances with a positive slope of $\alpha_{REF,BG}=1.25$ are observed. The resulting electron occupation numbers of the QD are given in white letters. (e) Conductance of the GNR depending on V_{BG} and V_{ref} with $V_{GNR}=0.5$ mV. (f) Absolute value of the GNR transconductance. Elevated conductance traces with a positive slope (dotted line) match perfectly with the Coulomb resonances in (d). Detection lines are even visible where the current is too small to be measured directly (see white arrows). Measurements shown in (a)-(c) were measured parallel to line (2) with an offset of $V_{BG}=48.166$ V. All measurements were recorded simultaneously.

to excited states providing additional transport channels. The excited state level spacing is obtained directly from this measurement (see white dotted line in Fig. 3(c)) resulting in Δ =0.9 meV.

Following Ref. [23] we can relate the level spacing with the length of the nanotube segment forming the quantum dot ($L_{\rm QD}$) by $\Delta{=}0.5$ eV/L(nm). This provides an estimate of $L_{\rm QD}\approx\!500$ nm which is in reasonable agreement with the device geometry and indicates that the QD extends over the entire nanotube segment defined by the metal source and drain contacts. Additionally, a similar length scale for the QD (150-235 nm) is obtained from the charging energy of 6-9 meV by EC=1.4 eV/L(nm) [23].

The independently measured back gate characteristic of the bilayer graphene nanoribbon is shown in Fig. 3(b). This low bias ($V_{\rm GNR}=0.5~{\rm mV}$) measurement highlights that transport can be tuned from the hole regime (left inset) into the so-called transport gap [24–31] starting at around $V_{\rm BG}{\approx}40~{\rm V}$. Within the gap region transport is governed by localized states resulting in sharp resonances of the conductance as shown in the right inset of Fig. 3(c). Interestingly, both carbon nanostructures have different doping levels. In contrast to the n-doped CNT we observe a significant p-doping of the GNR which is most likely due to atmospheric O_2 binding on the graphene edges [32]. Fig. 3(d) shows the conductance of the CNT in dependence of the BG voltage $V_{\rm BG}=13.63~{\rm V}+\Delta V_{\rm BG}$

and the reference voltage on the nanoribbon V_{REF^*} at $V_{CNT}{=}20$ mV, which demonstrates the gating effect of the GNR on the CNT QD. Lines of higher conductance can be attributed to Coulomb resonances of the CNT and from their slopes we extract a relative lever arm of $\alpha_{BG,GNR}{=}0.23$.

If both quantum devices are now operated simultaneously, the nanoribbon device can be used to detect individual charging events on the nanotube QD device. This phenomenon is shown in Fig. 4. The BG voltage is put to an offset (V_{BG} =48.166 V) such that (i) the CNT is in the Coulomb blockade regime and (ii) the conductance of the nanoribbon exhibits sharp and well-reproducible resonances. Figs. 4(a) and 4(b) show the simultaneously measured low-bias (V_{CNT} =0.5 meV) conductance through the nanotube G_{CNT} and the nanoribbon G_{GNR} as function of V_{REF} respectively. Similar to the inset of Fig. 3(a) G_{CNT} exhibits Coulomb peaks which are indicating single charging events in the CNT QD. In the simultaneously measured trace of the GNR we observe distinct steps in the conductance at the exact positions of the CNT QD charging events. The steps in conductance can measure up to 20 % of the total resonance amplitude and are due to the capacitive coupling of both nanostructures. Increasing the reference potential V_{REF} and decreasing V_{BG} both leads to a higher chemical potential in the QD and subsequently to a lower occupation number at every event. Consequently, the GNR resonance

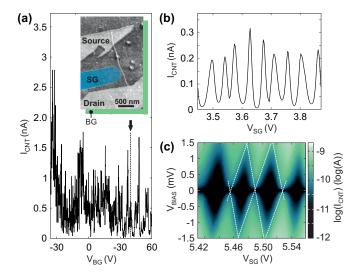


FIG. 5. (color online) (a) Back gate characteristics of a carbon nanotube contacted by two graphene flakes and a bias voltage of $V_{\rm BIAS}{=}1~\rm mV$. The inset shows a SFM image of the investigated device (see also Fig. 1(e)). (b) Current $I_{\rm CNT}$ through the nanotube as function of the voltage $V_{\rm SG}$ applied to a metal electrode placed roughly 50 nm from the nanotube (see blue area in the inset of panel (a)) for a fixed back gate voltage $V_{\rm BG}{=}35~\rm V$ and $V_{\rm BIAS}{=}0.3~\rm mV$. (d) Current $I_{\rm CNT}$ through the nanotube on a logarithmic scale as function of $V_{\rm BIAS}$ and $V_{\rm SG}$. $V_{\rm BG}$ is constant at 35.15 V.

shifts to lower values of V_{REF} giving rise to the unconventional shape of the charge detecting resonance in Fig. 4(b) as illustrated by the inset. By relating the step height to the noise level of our measurement system we achieve an estimate for the charge sensitivity with an upper limit of 10^{-3} e/ $\sqrt{\text{Hz}}$. In order to highlight the charge detection we further plot the transconductance i.e. the derivative of G_{GNR} with respect to V_{REF} as shown in Fig. 4(c). Each individual transconductance peak (dip) is very well aligned with the directly measured Coulomb peaks. For proving this more rigerously we performed measurements as function of both $V_{\rm REF}$ and $V_{\rm BG}$ and extracted relative lever arms. Corresponding charge stability diagrams are shown in Figs. 4(d) and 4(e). As anticipated, the evolution of Coulomb resonances of the CNT QD (shown in Fig. 4(d)) follows a constant positive slope with a relative lever arm of $\alpha_{\rm REF,BG} \approx 1.25$ and the occupation numbers (denoted in white letters) decrease with increasing V_{REF} and decreasing V_{BG}. For the charge detector the observed patterns in Fig. 4(e) show similarities to those of a multi-dot system. Further analysis of the transconductance dG_{CD}/dV_{REF} reveals at least three lines with negative slopes. Each of these lines can be attributed to different dot features in the graphene nanoribbon with relative lever arms of $\alpha_{\rm REF,BG}^{(1)}$ = -0.43, $\alpha_{\rm REF,BG}^{(2)}$ = -0.31 and $\alpha_{\rm REF,BG}^{(3)} = -0.12$ as indicated in Fig. 4(f). Additionally the plot exhibits clearly visible features (see white dotted line) with slopes and positions matching perfectly to those in Fig. 4(d). Consequently they can be associated with charging events in the CNT QD. Detection lines are even visible where the current through the dot is too low to be measured directly (see white arrows in Figs. 4(d) and (f)).

Finally, we discuss a carbon nanotube device where both metal leads (source and drain) are substituted by two graphene flakes. The device structure has been discussed earlier (see fabrication section and Fig. 1(e)) and a close up of the final device is shown as inset in Fig. 5(a). For measuring the conductance of the nanotube, Cr/Au metal contacts are deposited on each graphene flake. Fig. 5(a) shows the back gate dependence of the current I_{CNT} through the nanotube. As a first important observation we see that current flows from one graphene flake to the other through the carbon nanotube. Moreover, the absence of a V_{BG} -regime where the current is fully suppressed suggests the presence of a metallic nanotube. A similar measurement is shown in Fig. 5(b) where we apply a voltage to a metallic side gate V_{SG} which was deposited next to the nanotube as indicated by the blue area in the inset of Fig. 5(a). The current I_{CNT} exhibits periodic Coulomb oscillations. In Fig. 5(c) we plot the current through the nanotube I_{CNT} on a logarithmic scale as function of the bias voltage $V_{\rm BIAS}$ and $V_{\rm SG}$ for a fixed value of $V_{BG}=35.15$ V (see black arrow in Fig. 5(a)). The measurement exhibits clear diamond shaped patterns which can be attributed to Coulomb diamonds. From the addition energies of 1-1.5 meV we can estimate the quantum dot length of 0.9-1.5 μ m [23], which is in good agreement with the 1.2 μ m spacing between the two graphene leads. This leads to the conclusion that the quantum dot in the nanotube extends over the full length set by the distance of the graphene leads. Moreover, the observation of Coulomb blockade oscillations and peaks manifests the presence of tunneling barriers at the interface of both carbon allotropes. The observed tunneling rates are going down to the low GHz regime. In conclusion, an all carbon quantum circuit which not only consists of gate structures, charge detectors or quantum dots but also includes an entire contacting scheme, can be realized exclusively using carbon components.

In summary, we present the fabrication and characterization of carbon nanotube-graphene hybrid devices. We show an example of a structure where a nanotube quantum dot is capacitively coupled to a graphene nanoribbon. Sharp resonances in the graphene nanoribbon gate characteristics give rise to clear charge detection signals with an upper estimate for the charge sensitivity of 10^{-3} e/ $\sqrt{\text{Hz}}$. The presented charge detector is rather easy to fabricate and its sensitivity might be further improved by implementing it into a rf-circuit [33]. Furthermore, we study a device where both allotropes are electrically coupled. We find that the interface between the graphene and the carbon nanotube resembles tunneling barriers. Both results open the road to more sophisticated devices which are entirely fabricated out of carbon nanostructures and exploit the different advantages of these promising materials.

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